

**We Claim:**

1. A method for purifying a mixture comprising single-wall carbon nanotubes and amorphous carbon contaminate, said method comprising the steps of:
- 5 of:
- (a) heating said mixture under oxidizing conditions sufficient to remove the said amorphous carbon and
- (b) recovering a product comprising at least about 80% by weight of single-wall carbon nanotubes.
- 10 2. The method of claim 1 wherein said oxidizing conditions comprise an aqueous solution of an inorganic oxidant.
3. The method of claim 2 wherein said inorganic oxidant is selected from the group consisting of nitric acid, a mixture of sulfuric acid and hydrogen peroxide, potassium permanganate and mixtures thereof.
- 15 4. The method of claim 2 wherein said aqueous solution is heated to reflux.
5. The method of claim 2 additionally comprising the step of subjecting the oxidized product of step (b) to a saponification treatment.
6. The method of claim 5 wherein said saponification treatment
- 20 comprises contacting said product with a basic solution.
7. The method of a claim 6 wherein said basic solution comprises sodium hydroxide.
8. The method of claim 6 additionally comprising the step of neutralizing the saponified product with an acid.
- 25 9. The method of claim 8 wherein said acid is hydrochloric acid.
10. The method of claim 8 additionally comprising the step of recovering a solid product from the saponified, neutralized product.
11. The method of claim 10 wherein said product is recovered by a method selected from the group consisting of filtration, settling by gravity, chemical
- 30 flocculators, and liquid cycloning.

12 The method of claim 10 wherein said solid product is a paper-like two dimensional product.

13 The method of claim 12 additionally comprising the step of drying the product.

14. The method of claim 13 wherein said product is dried at about 850°C in a hydrogen gas atmosphere

15 The method of claim 1 wherein said product comprises at least about 90% by weight of single-wall carbon nanotubes.

16 The method of claim 1 wherein said product comprises at least about  
10 95% by weight of single-wall carbon nanotubes.

17 The method of claim 1 wherein said product comprises at least about 99% by weight of single-wall carbon nanotubes.

18 A method for producing tubular carbon molecules of about 5 to 500 nm in length, said method comprising the steps of:

15 (a) cutting single-wall nanotube containing-material to form a mixture of tubular carbon molecules having lengths in the range of 5-500 nm;

(b) isolating from said mixture of tubular carbon molecules a fraction of said molecules having substantially equal lengths.

20            19.    The method of claim 18 wherein said cutting single-wall nanotubes  
into tubular carbon molecules comprising the steps of:

(a) forming a substantially two-dimensional target containing single-wall nanotubes of lengths up to about one micron or more, and

(b) irradiating said target with a high energy beam of high mass ions.

25            20.    The method of claim 19 wherein a high energy beam is produced in  
a cyclotron and has an energy of from about 0.1 to about 10 GeV.

21. The method of claim 19 wherein said high mass ion has a mass of greater than about 150 AMU.

22. The method of claim 21 wherein said high mass ion is selected from  
30 the group consisting of gold, bismuth and uranium.

- 23 The method of claim of 22 wherein the high mass ion is Au<sup>-33</sup>
- 24 The method of claim 18 wherein said cutting single-wall nanotubes  
into tubular carbon molecules comprises the steps of:
- 5 (a) forming a suspension of single-wall nanotubes in a medium,  
(b) sonicating said suspension with acoustic energy
- 25 The method of claim 24 wherein said acoustic energy is produced  
by a device operating at 40 KHz and having an output of 20 W
- 26 The method of claim 18 wherein said cutting single-wall nanotubes  
into tubular carbon molecules comprises refluxing single wall nanotube material in  
10 concentrated HNO<sub>3</sub>.
- 27 The method of claim 19 further comprising the step of heating the  
tubular carbon molecules to form a hemispheric fullerene cap on at least one end  
thereof.
- 28 The method of claim 18 further comprising the step of reacting said  
15 tubular carbon molecules with a material which provides at the reaction conditions  
at least one substituent on at least one of said ends of said tubular carbon molecule.
- 29 The method of claim 26 further comprising the step of reacting said  
tubular carbon molecules with a material which provides at the reaction conditions  
at least one substituent on at least one of said ends of said tubular carbon molecule
- 20 30 The method of claim 28 or 29 wherein said substituent is selected  
from the group consisting of each may be independently selected from the group  
consisting of hydrogen; alkyl, acyl, aryl, aralkyl, halogen; substituted or  
unsubstituted thiol; unsubstituted or substituted amino; hydroxy, and OR' wherein  
R' is selected from the group consisting of hydrogen, alkyl, acyl, aryl aralkyl,  
25 unsubstituted or substituted amino; substituted or unsubstituted thiol, and halogen;  
and a linear or cyclic carbon chain optionally interrupted with one or more  
heteroatom, and optionally substituted with one or more =O, or =S, hydroxy, an  
aminoalkyl group, an amino acid, or a peptide of 2-8 amino acids.
- 30 31 A method for forming a macroscopic molecular array of tubular  
carbon molecules, said method comprising the steps of:

- (a) providing at least about  $10^6$  tubular carbon molecules of substantially similar length in the range of 50 to 500 nm;
- (b) introducing a linking moiety onto at least one end of said tubular carbon molecules,
- 5 (c) providing a substrate coated with a material to which said linking moiety will attach; and
- (d) contacting said tubular carbon molecules containing a linking moiety with said substrate

32. The method of claim 31 wherein said substrate is selected from the  
10 group consisting of gold, mercury and indium-tin-oxide.

33. The method of claim 32 wherein said linking moiety is selected from the group consisting of -S-, -S-(CH<sub>2</sub>)<sub>n</sub>-NH-, and -SiO<sub>3</sub>(CH<sub>2</sub>)<sub>3</sub>NH<sub>2</sub>

34. A method for forming a macroscopic molecular array of tubular carbon molecules, said method comprising the steps of:

- 15 (a) providing a nanoscale array of microwells on a substrate;
- (b) depositing a metal catalyst in each of said microwells; and
- (c) directing a stream of hydrocarbon or CO feedstock gas at said substrate under conditions that effect growth of single-wall carbon nanotubes from each microwell.

20 35. The method of claim 34 further comprising the step of applying an electric field in the vicinity of said substrate to assist in the alignment of said nanotubes growing from said microwells.

36. A method for forming a macroscopic molecular array of tubular carbon molecules, said method comprising the steps of:

- 25 (a) providing surface containing purified but entangled and relatively endless single-wall carbon nanotube material;
- (b) subjecting said surface to oxidizing conditions sufficient to cause short lengths of broken nanotubes to protrude up from said surface; and

- (c) applying an electric field to said surface to cause said nanotubes protruding from said surface to align in an orientation generally perpendicular to said surface and coalesce into an array by van der Waals interaction forces.

5           37     The method of claim 36 wherein said oxidizing conditions comprise heating said surface to about 500°C in an atmosphere of oxygen and CO<sub>2</sub>

          38     A method of forming a macroscopic molecular array of tubular carbon molecules, said method comprising the step of assembling subarrays of up to 10<sup>6</sup> single-wall carbon nanotubes into a composite array.

10           39     The method of claim 38 wherein all the subarrays have the same type of nanotubes.

          40     The method of claim 38 wherein the subarrays have different types of nanotubes.

          41     The method of claim 38 wherein the subarrays are made according  
15 to the method of any of claims 31, 34 or 36.

          42     A method for continuously growing macroscopic carbon fiber comprising at least about 10<sup>6</sup> single-wall nanotubes in generally parallel orientation, said method comprising the steps of:

- 20           (a)     providing a macroscopic molecular array of at least about 10<sup>6</sup> tubular carbon molecules in generally parallel orientation and having substantially similar lengths in the range of from about 50 to about 500 nanometers,
- (b)     removing the hemispheric fullerene cap from the upper ends of the tubular carbon molecules in said array;
- 25           (c)     contacting said upper ends of the tubular carbon molecules in said array with at least one catalytic metal;
- (d)     supplying a gaseous source of carbon to the end of said array while applying localized energy to the end of said array to heat said end to a temperature in the range of about 500° C to about 1300° C; and
- 30           (e)     continuously recovering the growing carbon fiber.

43 The method of claim 42 wherein said fullerene caps are removed by heating in an oxidative environment

44 The method of claim 43 wherein said oxidative environment comprises aqueous etching with nitric acid or gas phase etching at temperatures of  
5 about 500°C in an atmosphere of oxygen and CO<sub>2</sub>.

45 The method of claim 42 wherein said catalytic metal is selected from the group consisting of Group VIII transition metals, Group VI transition metals, metals of the lanthanide series, metals of the actinide series, and mixtures thereof.

46 The method of claim 45 wherein said catalytic metal is selected from  
10 the group consisting of Fe, Co, Ni, Ru, Rh, Pd, Os, Ir and Pt

47 The method of claim 46 wherein said catalytic metal is selected from the group consisting of Fe, Ni, and Co, and mixtures thereof.

48 The method of claim 45 wherein said catalytic metal is selected from the group consisting of Cr, Mo, and W.

15 49 The method of claim 42 wherein said catalytic metal is deposited in situ on each nanotube as a metal atom cluster.

50 The method of claim 49 wherein said metal atom cluster has from about 10 to about 200 metal atoms.

51 The method of claim 42 wherein said catalytic metal is deposited as  
20 preformed nanoparticles.

52 The method of claim 51 wherein said catalytic metal is Mo

53 The method of claim 42 wherein said catalytic metal is deposited in the form of a metal precursor selected from the group consisting of salts, oxides and complexes of said metal.

25 54 The method of claim 42 wherein said catalytic metal is deposited by evaporating metal atoms and allowing them to condense and coalesce on said open nanotube ends.

55 The method of claim 54 wherein said evaporation is effected by heating a wire or wires containing said catalytic metal.

56. The method of claim 54 wherein said evaporation is effected by molecular beam evaporation.

57. The method of claim 42 wherein gaseous source of carbon is selected from the group consisting of hydrocarbons and carbon monoxide.

5 58. The method of claim 57 wherein said hydrocarbon is selected from the group consisting of alkyls, acyls, aryls and aralkyl having 1 to 7 carbon atoms

59. The method of claim 58 wherein said hydrocarbon is methane, ethane, ethylene, acetylene, acetone, propane, propylene and mixtures thereof.

10 60. The method of claim 42 wherein said localized energy is provided by a laser beam.

61. The method of claim 42 wherein said localized energy is provided by a source selected from the group consisting of a microwave generator, an R-F coil and a solar concentrator.

15 62. The method of claim 42 wherein said end is heated to a temperature in the range of about 900°C to about 1100°C.

63. A composition of matter comprising at least about 80% by weight of single-wall carbon nanotubes.

64. The composition of claim 63 comprising at least about 90% by weight of single-wall carbon nanotubes.

20 65. The composition of claim 63 comprising at least about 95% by weight of single-wall carbon nanotubes.

66. The composition of claim 63 comprising at least about 99% by weight of single-wall carbon molecules.

25 67. A substantially two-dimensional article comprising at least about 80% by weight of single-wall carbon nanotubes.

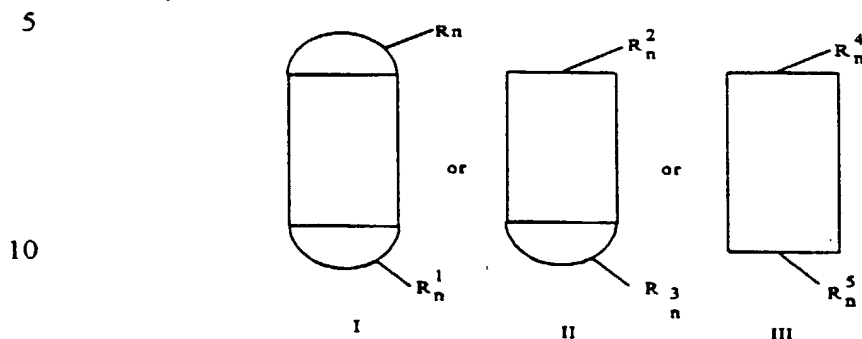
68. The article of claim 67 comprising at least about 90% by weight of single-wall nanotubes.

69. The article of claim 67 comprising at least about 95% by weight of single-wall nanotubes.

70. The article of claim 67 comprising at least about 99% by weight of single-wall nanotubes.

71. The article of claim 67 in the form of a paper-like material

72. A tubular carbon molecule having the following structure



where

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is a substantially defect-free cylindrical graphene sheet (optionally doped with noncarbon atoms) having from about  $10^2$  to  $10^6$  carbon atoms;

20

where



is a hemispheric fullerene cap having at least six pentagons and the remainder hexagons;

25

n

is a number from 0 to 30; and

$R, R^1, R^2, R^3, R^4,$  and  $R^5$

each may be independently selected from the group consisting of hydrogen; alkyl, acyl, aryl, aralkyl, halogen; substituted or

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unsubstituted thiol; unsubstituted or substituted amino, hydroxy, and OR' wherein R' is selected from the group consisting of hydrogen, alkyl, acyl, aryl, aralkyl, unsubstituted or substituted amino, substituted or unsubstituted thiol, and halogen; and a linear or cyclic carbon chain optionally interrupted with one or more heteroatom, and optionally substituted with one or more =O, or =S, hydroxy, an aminoalkyl group, an amino acid, or a peptide of 2-8 amino acids.

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73 The molecule of claim 72 wherein said graphene sheet has a configuration that corresponds to a (n,n) single-wall carbon nanotube.

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74. The molecule of claim 72 wherein said molecule has a length from about 5 to about 1000 nm.

75. The molecule of claim 74 wherein said molecule has a length of from about 5 to about 500 nm.

76 The molecule of claim 72 wherein n is 0 to 12

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77. The molecule of claim 72 further comprising at least one endohedral species.

78. The molecule of claim 77 wherein said endohedral species is selected from the group consisting of metal atoms, fullerene molecules, other small molecules and mixture thereof.

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79. The molecule of claim 78 comprising a (10,10) single-wall nanotube containing at least one endohedral species selected from the group consisting of C<sub>60</sub>, C<sub>70</sub>, or mixtures thereof.

80. The molecule of claim 79 wherein said C<sub>60</sub> or C<sub>70</sub> additionally contains an endohedral substituent selected from the group consisting of metal atoms and metal compounds.

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81. A macroscopic molecular array comprising at least about  $10^6$  single-wall carbon nanotubes in generally parallel orientation and having substantially similar lengths in the range of from about 5 to about 500 nanometers.
82. The array of claim 81 wherein said nanotubes are of the same type
- 5 83. The array of claim 82 wherein said nanotubes are of the (n,n) type
84. The array of claim 83 wherein said nanotubes are of the (10,10) type.
85. The array of claim 83 wherein said nanotubes are of the (m,n) type
86. The array of claim 81 wherein said nanotubes are of different types
- 10 87. The array of claim 81 further comprising a substrate attached to one end of said array and oriented substantially perpendicularly to the nanotubes in said array
88. The array of claim 87 wherein said substrate is a bucky paper surface.
- 15 89. The array of claim 87 wherein said substrate is a metal layer selected from the group consisting of gold, mercury and indium-tin-oxide.
90. The array of claim 86 wherein a central portion of nanotubes are of the (n,n) type and an outer portion of nanotubes are of the (m,n) type
- 20 91. A macroscopic carbon fiber comprising at least about  $10^6$  single-wall carbon nanotubes in generally parallel orientation.
92. The fiber of claim 91 comprising at least about  $10^9$  single-wall carbon nanotubes.
93. A composite fiber comprising a plurality of the fibers of claim 91
94. A molecular template array for growing continuous length carbon
- 25 fiber comprising a segment of the fiber of claim 91.
95. The fiber of claim 91 having a length of at least 1 millimeter.
96. The fiber of claim 91 wherein a substantial portion of said nanotubes are of the (n,n) type.
97. The fiber of claim 91 wherein all of said nanotubes are not of the
- 30 same type.

98. A composite article of manufacture comprising a matrix material selected from the group consisting of metals, polymers, ceramics and cermets, said matrix having embedded in at least a portion thereof a property enhancing amount of the carbon fibers of claim 91.

5 99. The composite article of claim 98 wherein said property is structural, mechanical electrical, chemical, optical, or biological.

100. A high voltage power transmission cable wherein at least one conductor comprises a continuous carbon fiber according to claim 96

101. The power transmission cable of claim 100 wherein both a central  
10 conductor and a coaxially disposed outer conductor are formed from said carbon fiber and an insulating layer is disposed therebetween.

102. The power transmission cable of claim 101 wherein said insulating layer is an air space.

103. The power transmission cable of claim 101 wherein said insulating  
15 layer comprises a material selected from the group consisting of insulating carbon fiber made from carbon nanotubes of the (m,n) type and insulating BN fiber made from hexaboronitride nanotubes or mixtures thereof.

104. A solar cell for converting broad spectrum light energy into electrical current comprising a molecular array according to claim 81 as the photon collector

20 105. The solar cell of claim 104 additionally comprising a photoactive dye coupled to the upper ends of the nanotubes in said array.

106. A bistable, nonvolatile memory bit comprising the endohedrally-loaded tubular carbon molecule of claim 77.

107. The memory bit of claim 106 wherein the tubular carbon molecule  
25 is formed from a (10,10) type nanotube and the endohedral species is a C<sub>60</sub> or C<sub>70</sub> fullerene molecule.

108. A bistable, nonvolatile memory device comprising the memory bit of claim 106, means for writing to said bit and means for reading said bit.

109. The memory device of claim 108 wherein said means for writing  
30 comprises a nanocircuit element adapted to direct a voltage pulse of positive or

negative polarity at said bit to cause said endohedral species to move from a first end to a second end of said bit.

110 The memory device of claim 108 wherein said means for reading said bit comprises

- 5 (a) a first nanocircuit element adapted to be biased at a first voltage ( $V_{Read}$ ) and spaced from a read end of said bit to form a first gap therebetween, and  
(b) a second nanocircuit element adapted to be biased to ground voltage ( $V_G$ ) and spaced from said read end of said bit to form a second gap, whereby the presence of said endohedral species is unambiguously determined by the presence  
10 of current tunneling across said first and second gaps.

111 A microporous anode for an electrochemical cell comprising a molecular array according to claim 81.

112. A lithium ion secondary battery comprising the anode of claim 111, a cathode comprising  $\text{LiCoO}_2$  and an aprotic organic electrolyte wherein a fullerene  
15 intercalating compound (FIC) of lithium forms at the anode under charging conditions.

(113) An apparatus for forming a continuous macroscopic carbon fiber from a macroscopic molecular template array comprising at least about  $10^6$  single-wall carbon nanotubes having a catalytic metal deposited on the open ends of said  
20 nanotubes, said apparatus comprising:

- (a) means for locally heating only said open ends of said nanotubes in said template array in a growth and annealing zone to a temperature in the range of about  $500^\circ\text{C}$  to about  $1300^\circ\text{C}$ ;  
(b) means for supplying a carbon-containing feedstock gas to the  
25 growth and annealing zone immediately adjacent said heated open ends of said nanotubes in said template array; and  
(c) means for continuously removing growing carbon fiber from said growth and annealing zone while maintaining the growing open end of said fiber in said growth and annealing zone.

114. The apparatus of claim 113 wherein said means for locally heating comprises a laser

115. The apparatus of claim 113 enclosed in a growth chamber maintained at a vacuum by evacuation means.

5 116. The apparatus of claim 115 further comprising a vacuum feed lock zone through which said continuously produced carbon fiber is passed and a take-up roll at atmospheric pressure.

117. A composite material comprising:

- 10 (a) a matrix; and  
(b) a carbon nanotube material embedded within said matrix.

118. The composite material of claim 117, wherein said matrix comprises a polymer.

119. The composite material of claim 118, wherein said polymer comprises a thermosetting polymer.

15 120. The composite material of claim 119, wherein said thermosetting polymer is selected from the group consisting of phthalic/maelic type polyesters, vinyl esters, epoxies, phenolics, cyanates, bismaleimides, and nadic end-capped polyimides.

20 121. The composite material of claim 118, wherein said polymer comprises a thermoplastic polymer.

122. The composite material of claim 121, wherein said thermoplastic polymer is selected from the group consisting of polysulfones, polyamides, polycarbonates, polyphenylene oxides, polysulfides, polyether ether ketone, polyether sulfones, polyamide-imides, polyetherimides, polyimides, polyarylates,  
25 and liquid crystalline polyesters.

123. The composite material of claim 117, wherein said matrix comprises a metal.

124. The composite material of claim 117, wherein said matrix comprises a ceramic.

125 The composite material of claim 117, wherein said matrix comprises a cermet

126 The composite material of claim 117, wherein said carbon nanotube material comprises tubular carbon nanotube molecules

5 127 The composite material of claim 117, wherein said carbon nanotube material comprises ropes up to about  $10^3$  SWNTs.

128 The composite material of claim 117, wherein said carbon nanotube material comprises fibers of greater than  $10^4$  SWNTs.

10 129 The composite material of claim 126, 127, or 128, further comprising an additional fibrous material.

130 The composite material of claim 126, 127, or 128, wherein said carbon nanotube material is modified to interact with said matrix material

131 A method for producing a composite material containing carbon nanotube material comprising:

- 15 (a) preparing a matrix material precursor;  
(b) combining a carbon nanotube material with said matrix material precursor; and  
(c) forming said composite material.

20 132 The method of claim 131, wherein said carbon nanotube material is combined with said matrix material precursor before said step of forming.

133 The method of claim 131, wherein said carbon nanotube material is combined with said matrix material precursor during said step of forming.

25 134 The method of claim 131, wherein said carbon nanotube material is combined with said matrix material precursor immediately after said step of forming.

135 The method of claim 131, wherein said matrix material precursor is caused to flow around a pre-formed arrangement of said carbon nanotube material.

136 A method of producing a composite material containing carbon nanotube material comprising:

- 30 (a) preparing an assembly of a fibrous material;

(b) adding said carbon nanotube material to said fibrous material, and

(c) adding a matrix material precursor to said carbon nanotube material and said fibrous material.

137 The method of claim 136, wherein said fibrous materials are  
5 arranged in a two-dimensional sheet, and some portion of the said carbon nanotube material is oriented in a direction other than parallel to said sheet.

138. The method of claim 131 or 136 wherein said carbon nanotube material comprises tubular carbon nanotube molecules.

139 The method of claim 131 or 136, wherein said carbon nanotube  
10 material comprises ropes of up to about  $10^3$  SWNTs.

140 The method of claim 131 or 136, wherein said carbon nanotube material comprises fibers of greater than  $10^6$  SWNTs.

141. A three-dimensional structure that self-assembles from derivatized single-wall carbon nanotube molecules comprising:

15 a plurality of multifunctional single-wall carbon nanotubes assembled into said three-dimensional structure.

142. The three-dimensional structure of claim 141, wherein said single-wall carbon nanotubes have multifunctional derivatives on their end caps

143. The three-dimensional structure of claim 141, wherein said single-  
20 wall carbon nanotubes have multifunctional derivatives at multiple locations on said single-wall carbon nanotubes.

144. The three-dimensional structure of claim 141, wherein said single-wall carbon nanotubes are assembled as a result of van der Waals attractions.

145. A three-dimensional structure of claim 141, which has  
25 electromagnetic properties.

146 The three-dimensional structure of claim 145, wherein said electromagnetic properties are determined by a functionally-specific agent.

147 A three-dimensional structure of claim 141, which is symmetrical.

148 A three-dimensional structure of claim 141, which is not  
30 symmetrical.

149 A three-dimensional structure of claim 141, which has biological properties.

150 A three-dimensional structure of claim 149, which operates as a catalyst for biochemical reactions.

5 151. A three-dimensional structure of claim 149, which interacts with living tissue

152. A three-dimensional structure of claim 149, which serves as an agent for interaction with functions of a biological system.

153. A light harvesting antenna comprising:  
10 at least one single-wall carbon nanotube conductive element, said at least one nanotube having a length selected relative to a desired current level and a desired voltage level.

154. The light harvesting antenna of claim 153, wherein said at least one single-wall carbon nanotube forms a Schottky barrier.

15 155. An array of light harvesting antennas of claim 153

156. The array of light harvesting antennas of claim 155, wherein said array is formed by self-assembly.

157 A molecular electronic component comprising at least one single-wall carbon nanotube.

20 158. The molecular electronic component of claim 157, wherein said molecular electronic component is a bridge circuit for providing full wave rectification, said bridge circuit comprising:

four single-wall carbon nanotubes, each of said four single-wall carbon nanotubes forming one edge of a square and linked to two of four  
25 buckyballs, each of said four buckyballs located at a corner of said square.

159. The bridge circuit of claim 158, wherein said buckyballs and single-wall carbon nanotubes are derivitized to include functionally specific linking agents.

160. A molecular electronic component of claim 157, which is a fullerene diode.



- 161 A nanoscale manipulator comprising at least one single-wall carbon nanotube.
162. The nanoscale manipulator of claim 161, which is nanoforceps